

A Review on Waste Management of Radioactivity Materials and its Economic Effects

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Abstract

Natural radioactivity is omnipresent on earth: natural radio nuclides are abundant in all oceans, in the earth's crust and in the atmosphere. Addition to natural resources, many activities dealing with radioactive materials produce nuclear wastes. These include civilian nuclear power reactors, defense nuclear programs (nuclear weapons production and related R&D), industrial and institutional activities and isotopic applications. To minimize the potential adverse health impacts to people during the entire lifetime of the radionuclide involved, nuclear waste must be carefully and properly managed. The scope of nuclear waste management encompasses generation, processing (treatment and packaging), storage, transport and disposal. In recent years, nuclear waste management in ceramics (e.g., synthetic rocks) has been one of the best options for immobilization of dangerous wastes and many research articles have approved the ability of these materials. In this article, the main synthetic rocks (Hollandite, Zirconolite, Perovskite, Pyrochlore and Brannerite) and process technologies for construction of them including sintering, HotIsostatic Pressing (HIP) in argon and Cold-Crucible Induction Melting have been discussed and applications of this type of waste management on immobilization of dangerous wastes are explained.

Keywords: Health Effects; Immobilization; Nuclear Waste Management; Radio nuclides; Synthetic Rocks

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1. Introduction

The term "radioactive waste" covers a wide variety of materials ranging from wastes that can be put safely into a dustbin to items that need remote handling, heavy shielding and cooling to be managed safely. To be clear, radioactive waste is considered in different categories [1-3]. These are:

a) Very Low-Level Waste (VLLW) of uranium mill tailings which are by-product material from the rough processing of uranium-bearing ore. VLLW is a subset of LLW and falls into two distinct categories: (1) Low volume VLLW

("dustbin disposal") that can be safely disposed of to an unspecified destination with municipal, commercial or industrial waste (the radioactive risk from such material is low enough that controls on disposal of this material, after removal from the premises where the wastes arose, are not necessary) and (2) High volume VLLW ("bulk disposal") that can be disposed of to specified landfill sites (after the waste is removed from its site of origin, it will be subject to controls on its disposal, which will be specified by the environmental regulators) [4].

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b) Low-Level Waste (LLW), is generated from hospital and industries as well as the nuclear fuel cycle. To reduce its volume, it is often compacted or incinerated before disposal. Most LLW today arises from the operation of nuclear power stations and nuclear fuel reprocessing facilities, as well as the decommissioning and clean up nuclear sites. Operational LLW is principally lightly contaminated miscellaneous waste arising from maintenance and monitoring, such as plastic, paper and metal. LLW from the decommissioning is mainly soil, building material and metal plants and equipments.

c) Intermediate Level Waste (ILW), contains higher amount of radioactivity and in some cases requires shielding. ILW includes resins, chemical sludge and metal reactor fuel cladding, as well as contaminated materials. This waste has radioactivity levels that are higher than LLW, but do not generate enough heat to require special storage or disposal facilities [5, 6]. However, like other radioactive wastes, it still needs to be contained to protect workers from the radiation.

d) High Level Waste (HLW), contains fission products and transuranic elements generated in the reactor core. This waste is radioactive enough for the decay heat to significantly increase its temperature and the temperature of its surroundings. This means heat generation should be taken into account when storage and disposal facilities are designed [7]. HLW includes (1) the liquid residue that contains most of the radioactivity from the reprocessing of spent nuclear fuels and (2) spent fuel (if it is declared a waste) or any other waste with similar radiological characteristics.

e) Transuranic Waste (TRUW), which is contaminated with alpha-emitting transuranic radionuclide with half-life greater than 20 years and concentration greater than 100 nCi/g (3.7 MBq/kg), excluding high level waste [8, 9].

The Nuclear Waste Management (NWM) plays a key role in the nuclear power industry and involves short-term management which deals with immediate treatment of the waste (Vitrification [10], Ion-exchange [11] and Synroc [12]) and long-term management including storage [13], disposal and transformation of waste into a non-toxic form [14, 15].

In recent years, NWM in ceramics has been one of the best options for immobilization of dangerous wastes and many research articles have approved the ability of these materials. Synthetic rock or Synroc may be the most well-known ceramic technology for the immobilization of nuclear wastes [16]. For this reason, the main synthetic rocks (Hollandite, Zirconolite, Perovskite, Pyrochlore and Brannerite) and process

technologies for construction of them including sintering, Hot-Isostatic Pressing in argon and Cold-Crucible Induction Melting have been discussed here, and applications of this type of waste management on immobilization of dangerous wastes are explained.

2. Immobilization in ceramics

The management of radioactive waste produced in nuclear power plants is a crucial problem facing developed countries [17]. The long history of research on this topic has shown that crystalline ceramics are among the most promising materials for use as matrices for the immobilization and/or transmutation of nuclear waste [18].

Due to the ease of waste immobilization in glass matrices (by melting and casting) and the high structural flexibility offered by such disordered materials, they are currently used (e.g. aluminosilicates glasses) to incorporate the broad range of high level radioactive wastes fission products plus minor actinides- originating from the reprocessing of commercial or defense spent fuels. Nevertheless, because of both the higher chemical durability and the higher capacity of several single phase ceramic matrices such as zirconates, titanates and phosphates to incorporate specific nuclear wastes (corresponding to either specific element or to a family of elements with similar chemical properties), ceramic waste forms have been proposed for the immobilization of long-lived radionuclides such as excess weapons plutonium and minor actinides [19].

Colloids are known to influence the migration behavior of long-lived radionuclides in the aquatic environment. These could be intrinsic colloids generated following the hydrolysis of higher valent actinides, e.g., Pu (IV), Am (III), etc or pseudo-colloids consisting of radionuclides sorbed on colloids formed by the weathering of surrounding rocks of water bodies. The common colloids present in the natural water are the silica, alumina, aluminosilicates, and iron oxides, e.g., hematite, magnetite, etc. Modeling of the sorption behavior of radionuclides by these colloids is carrying out using the surface complexation models. As natural colloids are complex in terms of their surface sites, crystalline state, and particle size distribution, modeling is carried out with model systems, which are simple and have well defined surface sites and structure as well as particle size.

Aluminosilicates form are the major components of rocks, clays and minerals present in the vicinity of ground water. Owing to their high chemical, thermal and radiation resistivity, as well as high retention capacity for metal ions, they are proposed as the best backfill materials (viz., bentonite, montmorillonite, etc.) for deep geological repository. However, owing to their large surface

area and high activity for sorption, aluminosilicates colloids can play an important role in the migration of radionuclides in aquatic environments [20].

The science of immobilizing of materials for nuclear applications has been an important topic for several years and many different studies have been performed and reported on this topic. The interest has varied from determining physical properties of many types of materials to modeling long-term performances of materials suggested for immobilization hosts [21].

3. Synroc

Perhaps the most well-known ceramic technology for the immobilization of nuclear wastes is synroc [16]. This material is a type of "synthetic rock", invented in 1978 by the late Professor Tedd Ringwood of the Australian National University [22]. The primary objective of the Synroc strategy for HLW management is to provide a waste form which has a much greater resistance to leaching by ground waters than borosilicate glass and which is capable of maintaining its integrity in suitable geological environments for periods exceeding one million years [23]. Synroc can take various forms depending on its specific use and can be tailored to immobilize particular components in the HLW. The main minerals in synroc are Hollandite, Zirconolite, Perovskite, Pyrochlore and Brannerite which have been explained in the next [24].

3.1. Hollandite

The Hollandite group of minerals has the general formula $A_xB_yC_{8-y}O_{16}$. In the crystal structure of Synroc-type Hollandite, the B and C cations are surrounded by octahedral configuration of oxygen. Each of these (B,C) O_6 octahedral share two edges to form paired chains running parallel to the C-axis. These chains are corner-linked to neighboring paired chains to form a three-dimensional framework with tunnels running parallel to the C-axis. The larger cations are located in these tunnels. In Synroc-type Hollandite, the A position is occupied by Cs/Rb and Ba, the B position by Al and Ti^{3+} , and the C position by Ti^{4+} [10, 25].

Hollandite is one of the major phases of Synroc, a multiphase ceramic matrix also containing Zirconolite and Perovskite phases that is being considered as a matrix for immobilizing Cs resulting from enhanced separation of fission product solutions from spent fuel reprocessing. The advantage of this material is that it also incorporates Ba, a decay element of Cs. It also exhibits good thermal conductivity, an advantage due to radiogenic heat produced by decay of ^{137}Cs and ^{134}Cs , as well as good radiation resistance [25].

3.2. Pyrochlore

Pyrochlore is a derivative of the fluorite structure and has the general formula $A_2B_2X_6Y$, in which A and B are 8- and 6- coordinated cation sites, and X and Y are 4- coordinated anion sites. In general, Na, Ca, Y, lanthanides and actinides can be incorporated on the A site, and Ti, Zr, Nb, Hf, Ta, Sn and W can be incorporated on the B site. Recent studies have revealed that even fully amorphous titanite pyrochlore samples (^{238}Pu - based) have very similar dissolution rates as those of the corresponding crystalline samples [26], showing that the chemical durability of the pyrochlore structure is sustained even when it is fully amorphous due to radiation damage. Pyrochlore-rich ceramics are among the specific compositions developed initially for immobilization of surplus impure plutonium using sintering as the consolidation process and have since been tailored to suit various actinide-rich waste streams. Nevertheless, all these waste forms contain sufficient amount of targeted additional phases to allow compositional flexibility to deal with variations in waste chemistry and waste precursor ratio [27].

3.3. Zirconolite

Zirconolite (nominally $(CaZr_xTi_{3-x})O_7$ with $0.83 \leq x \leq 1.36$ has a monoclinic layered structure (C_2/c space group) referred as Zirconolite-2M polytype consisting of alternate planes containing either Ti^{4+} ions or both Ca^{2+} and Zr^{4+} ions [28]. Calcium and Zirconium ions are ordered within the planes in 8- and 7- fold coordinate sites, two of which ($Ti(1)$ and $Ti(3)$ sites) are 6- fold coordinated whereas the third ($Ti(2)$ site) half- occupied, is 5- fold coordinated. Zirconolite is well known for its excellent capacity to incorporate actinides and lanthanides into the Ca and Zr sites of its structure [19].

Zirconolite was the most durable phase design for the incorporation of actinides and lanthanides in synroc phase assemblages developed in Australia in 1980s for immobilization of unpartitioned Purex-type HLW produces by reprocessing of the spent fuels. More recently Zirconolite-rich and Zirconolite single phase ceramics were also envisaged as specific matrix for Pu-rich waste and minor actinide immobilization [29,30]. The long term performance of Zirconolite as actinide waste form are confirmed by the existence of very old natural samples which have incorporated and retained large amount of UO_2 and ThO_2 in the spite of metamictization phenomena due to radiation damages from alpha-decay. Moreover, recent results showed that amorphization produced by external irradiation (Au^{2+} ions) did not significantly affect the dissolution rate of Zirconolite [31].

3.4. Brannerite

Brannerite (UTi_2O_6), which exists naturally in many uranium ore bodies, has attracted recent attention as a minor phase in the ceramic formulation designed to immobilize surplus plutonium. It belongs to the monoclinic crystal system with space group $C2/m$, and both U and Ti occupy distorted octahedral coordination polyhedral. The mineralogy and hydrometallurgy of brannerite in some ore deposits have been extensively studied. Several important observations have been made from the studies: (1) brannerite is more resistant than uraninite in to dissolution in sulphuric acid, (2) from the compositional and leach studies, sufficient evidence exists to support the presence of coffinite ($U[SiO_4]_{1-x}[OH]_{4x}$) as an alteration product inter growing with brannerite and (3) leach pit formation is initial, rarecontrolling step in brannerite dissolution regardless of the leach conditions [32-34].

3.5. Perovskite

A perovskite is any material with the same type of crystal structure as calcium titanium oxide ($CaTiO_3$), known as the perovskite structure, or $^{XII}A^{2+VI}B^{4+}X_3$ with the oxygen in the face centers [35]. The general chemical formula for perovskite compounds is ABX_3 , where 'A' and 'B' are two cations of very different sizes, and X is an anion that bonds to both. The 'A' atoms are larger than the 'B' atoms. The ideal cubic-symmetry structure has the B cation in 6fold coordination, surrounded by an octahedron of anions, and the A cation in 12-fold cuboctahedral coordination. Perovskite, nominally $CaTiO_3$, developed for immobilization of Sr, lanthanides and actinides [36].

4. Synroc processing

Parallel to the synroc formulation development, advanced synroc processing technologies, including sintering, cold crucible induction melting (CCIM) and hot isostatic pressing (HIP), have also been extensively studied.

4.1. Sintering

Sintering consists of pressing a pellet generally somewhat larger than the desired size and sintering it in the furnace to an appropriate temperature, via a carefully controlled heating/cooling cycle, under the desired atmosphere. Sintering is widely used in the nuclear industry for the production of MOX fuel pellets. To sinter a binder and lubricant must be added to press uniform "green" pellets, and the feed powder must be ground fine (≈ 10 microns) and mixed to increase activity. This increased reactivity will allow the pellets to be sintered without cracking or unpredictable dimensional changes and will avoid sticking between sintered pellets and

furnace equipment provide appropriate setter plates are used [1].

4.2. Hot Isostatic Pressing

A HIP consists of a pressure vessel surrounding a resistance-heated furnace. In hot isostatic pressing, a stainless steel can be filled with calcined radioactive waste feed and is then evacuated at temperature up to $500^{\circ}C$ and sealed. The can is placed into a furnace and the HIP vessel is closed and pressurized using argon gas. The pressure is applied isostatically via the dense high pressure argon, which is also an extremely efficient conductor of heat. This combined effect consolidates and immobilizes the waste into a dense isotropic monolithic block sealed within the can. HIPs are used in the nuclear industry and are installed in hot-shell facilities [27].

At HIPing conditions (high temperature and high pressure), the waste form and the stainless steel container used for containment of the waste form may potentially interact, which may in turn result in some detrimental effects on the quality of the waste form [37].

4.3. Cold-Crucible Induction Melting

Synroc can be formed by melting oxides into a glass composite material. Glass ceramics (crystalline) are made by melting to form a viscous glass, which is then crystallized on cooling or in a separate crystallizing heat treatment operation. Controlled crystallization can be used to desired phases rather than uncontrolled devitrification of a vitreous waste form. Uncontrolled crystallization can lead to potential stress and cracking or reduced durability. With controlled crystallization, there is a potential for higher waste loadings, and improved mechanical and chemical properties compared to the parent glass [38].

5. Immobilization in Synroc (Applications)

5.1. Immobilization of Cs/Sr

^{137}Cs isotopes approximately represent 14% of all cesium isotopes in UO_2 spent fuel and selective separation methods using functionalized calixarene molecules are under

development to isolate cesium from HLW. As transmutation of Cs cannot be envisaged without isotopic separation, it is proposed to immobilize all cesium isotopes in ceramic matrices [36].

Important factors in determining the applicability of using Synroc for immobilization of Cs and Sr include the potential waste loading (to reduce storage space requirements), the chemical durability (to reduce leach resistance as compared to borosilicate glass) and the waste form flexibility (to allow for feed/process variations). Much research has been completed in the past 20 years

on the development of Synroc for various types of Cs and Sr waste streams [1].

5.2. Immobilization of Pu-Rich Wastes

Zirconolite-Rich Synroc is a ceramic host for the immobilization option for the final disposition of weapon grade Pu [30]. Some of the Pu considered for immobilization contains significant impurities whose role in the waste form chemistry and processing must be understood.

The incorporation of pure Pu in Zirconolite utilizing processing atmospheres ranging from oxidizing to reducing is reasonably well understood on the basis of detail compositional, x-ray absorption and x-ray diffraction data. The Pu can enter the Ca site at levels up to ~ 0.5 formula unit (~ 30 wt% PuO₂), in either the +3 or +4 states, however only Pu⁴⁺ can enter the Zr site and then only at levels up to ~ 0.15 formula unit. At higher levels of Pu, mixtures of different Zirconolite structures form, sometimes together with pyrochlore, and these phases are distributed on a sub-micron scale, depending on the firing temperature [21].

5.3. Immobilization of actinides in phosphate matrices

In the field of immobilization of high-activity-level and long-life radwaste for a deep underground repository, several phosphate matrices have proposed to delay the release of actinides in the near-field of such disposal. Among them, thorium phosphate-diphosphate (TPD), monazite/brabantite, britholites, and TPD-monazite composites have been extensively studied. These materials could be potential candidates for the immobilization of actinides coming from an advanced reprocessing of spent fuel or for the final disposal of the excess plutonium from dismantled nuclear weapons [39].

6. Conclusion

Nuclear energy sustainability will depend on the actual capability of reducing the inventory and long-term radiotoxicity of nuclear waste, mainly dominated by the amount of transuranic isotopes remaining on the spent fuel. The long history of research on the management of radioactive waste produced in nuclear power plants, has shown that crystalline ceramics are among the most promising materials for use as matrices for the immobilization and/or transmutation of nuclear waste. Perhaps the most well-known ceramic technology for this purpose is synroc. Hence, the main topic of this article devoted to this matter.

The primary objective of the Synroc strategy for HLW management is to provide a waste form which has a much greater resistance to leaching by ground

waters than borosilicate glass and which is capable of maintaining its integrity in suitable geological environments for periods exceeding one million years. The main minerals in synroc are Hollandite, Zirconolite, Perovskite, Pyrochlore and Brannerite. In this study, the main synthetic rocks, Hollandite, Zirconolite, Perovskite, Pyrochlore and Brannerite, and their structures were introduced. Then, process technologies for construction of synroc including sintering, Hot-Isostatic Pressing (HIP) in argon and Cold-Crucible Induction Melting had been discussed and capability of this type of waste management on immobilization of dangerous wastes (Cs/Sr, Pu, actinides, plutonium, etc) were explained.

From the examined literature, it is clear that the safety level of nuclear power in several international locations has been successfully achieved. Also, the risks associated with nuclear power plants are much less than those due to any other available energy sources. Nevertheless, at present, there is a strong need to review educational requirements for under and post-graduate studies at universities to provide applied engineering skills required to design, build and operate simulated nuclear systems. In addition, national and international R&D programs are ought to be developed through the international forums to ensure the highest safety and security measures in nuclear power plants and waste management.

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